



5 ORGANIC LIGHT EMITTING DIODE (OLED) AND METHOD FOR THE
PRODUCTION THEREOF

RELATED APPLICATIONS

This is a U.S. national stage of International
application No. PCT/DE02/04346 filed 27 November, 2002.

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This patent application claims priority of German
patent application no. 10164016.1 filed 28 December
2001, the subject matter of which is hereby
incorporated by reference.

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FIELD OF THE INVENTION

The invention relates to an organic light emitting
diode (OLED for short) with at least one polymer layer
~~in accordance with the preamble of patent claim 1.~~

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BACKGROUND OF THE INVENTION

Organic light emitting diodes exhibit a series of
advantages which make them attractive for use in
optoelectronics. These include the availability of many
25 emission colors, low threshold voltages, fast
switchability, small thickness and the possibility of
using flexible substrates. Typical areas of application
of OLEDs are pixelated display instruments and large-
area elements for illumination purposes.

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One important development goal for OLEDs is to increase
the luminous efficiency and, associated with this, to
reduce the power consumption. This is of importance
particularly for portable electronic devices, in which
35 only limited energy resources are available.

The majority of the previous developments for
maximizing the luminous efficiency have been concerned

with increasing the internal quantum efficiency. The latter is defined as the ratio between the photons generated in the diode to the injected electrons. New materials with improved luminescence properties, 5 optimized layer sequences or better adapted electrode materials have contributed to increasing the internal quantum efficiency in recent years.

A different approach for increasing the luminous 10 efficiency consists in improving the extraction efficiency. Extraction efficiency is understood as the probability with which a photon generated in the emission zone can be coupled out of the diode and thus ejected. Coupling-out losses arise as a result of 15 absorption or as a result of waveguidance in one of the layers. Waveguidance is caused by total reflection at the interface of two layers with different refractive indexes. In the case of planar interfaces, the angle between incident and reflected beam does not change 20 apart from the sign. Accordingly, once a photon has been subjected to total reflection, it remains confined in the corresponding layer and cannot be extracted.

The representation of Figure 1 illustrates the process 25 of waveguidance in the layers of an organic light emitting diode:

A transparent electrode 2 (generally indium tin oxide, ITO for short) is situated on a substrate 3. At least 30 one organic layer 1 is deposited on said electrode, said layer being followed by an electrode 5 (e.g. a cathode). Photons are generated by emitters 6 in the organic layer 1. Only photons which do not remain in one of the layers 1-3 and 5 as a result of waveguidance 35 are extracted. The line IV shows the path of an extracted photon by way of example.

Depending on the layer thickness and refractive index of the individual layers, wave-guiding effects, indicated by the lines I, II and III in Figure 1, may occur in the organic layer, in the transparent electrode and in the substrate, respectively. Since the layer thicknesses of the organic layers and of the transparent electrode are in the region of the light wavelength or less than that, discrete optical modes I, II, corresponding respectively to lines I, II mentioned above, form in these layers, ~~while a mode as~~ a single stationary optical wave with a distinct wavelength. A continuum of modes III, corresponding to line III mentioned above, is present in the substrate as a plurality of optical waves with different wavelengths, and traditional geometric optics can be used here to calculate and predict the behavior of the continuum of modes. Therefore, a distinction is made between layer modes and substrate modes.

If the formation of layer modes can be disturbed and the undesirable waveguidance can thus be minimized, the extraction efficiency is increased and so, too, is the luminous efficiency.

The optical modes formed in a thin planar layer are dependent not only on the wavelength but essentially on the layer thickness and the refractive index of the layer.

A change in the refractive index and thus the disturbance of the wave-guiding effects can be achieved by means of inhomogeneities in the organic layer if the inhomogeneities have a different refractive index than the layer matrix.

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To date, the approaches a) to c) briefly outlined below have been proposed for suppressing wave-guiding effects, in which case a) and b) aim at changing the

refractive index and c) utilizes a change in the layer thickness:

a) Dispersion of nanoparticles in one of the organic
5 layers of the OLED (S.A. Carter et al., Enhanced
luminance in polymer composite light emitting devices,
Appl. Phys. Lett. 71(9), p. 1145, 1997)

In this case, particles of TiO_2 , SiO_2 or Al_2O_3 having a
size of 30 to 80 nm are embedded in the polymeric
10 emitter material MEH-PPV.

This method is associated with the following
difficulties:

It is technically very difficult to disperse
15 nanoparticles uniformly in a solvent in which polymers
have already been dispersed or dissolved. The
consequence of poorly dispersed nanoparticles is an
inhomogeneous emission of the LED layer which contains
said nanoparticles, for which reason diodes produced in
20 such a way are not suitable for use in display
instruments.

The oxidic nanoparticles proposed can lead to a
degradation of the active layer through oxidation.
Accordingly, a significantly shorter service life has
25 been observed in the case of the diodes with
nanoparticles than in the case of the reference diodes.

b) Densely packed SiO_2 microspheres (T. Yamasaki,
K. Sumioka, T. Tsutsui: Organic light-emitting device
30 with an ordered monolayer of silica microspheres as a
scattering medium, Appl. Phys. Lett. 76(10), p. 1243,
2000).

In this case, monolayers of densely packed SiO_2 spheres
with a diameter of 550 nm are used as scattering
35 centers. In this case, the spheres are applied on the
substrate beside the ITO anode tracks. This makes it
possible to suppress waveguidance in the organic layers

and in the glass. An increase in the coupling-out efficiency was observed.

5 This method is associated with the following difficulties:

The application of densely packed sphere areas can be realized only with a very high effort. Moreover, a large-area application of such areas of several square centimeters has not yet been realized hitherto. The
10 scattering centers are situated outside the active diode volume. Accordingly, only a small part of the substrate surface can be utilized for light generation. Furthermore, an inhomogeneous luminance results.

15 The periodic structure of the dense sphere packing means that the scattering efficiency is highly wavelength-selective. Accordingly, a generally undesirable lateral color profile occurs.

20 c) Corrugated (wave-shaped) organic layers (J.M. Lupton et al., Bragg scattering from periodically microstructured light emitting diodes, Appl. Phys. Lett. 77(21), p. 3340, 2000).

In this case, a polymeric LED is applied on a
25 one-dimensionally periodic structure with a period of 388 nm and depths of 10-100 nm. The structure acts as a Bragg reflector and again leads to scattering of optical modes in the emitter material.

30 This method is associated with the following difficulties:

The periodicity of the structure leads to a high degree of angular dispersion. A 15 nm thin gold layer was used as the anode, said gold layer already having a high
35 degree of absorption despite the small layer thickness. Transferring the corrugation to the transparent ITO that is otherwise customary as a standard anode is

difficult to realize on account of the larger ITO layer thicknesses and high process temperatures.

Furthermore, there are diverse approaches for better
5 coupling-out of the substrate modes. However, since these methods cannot prevent the formation of optical modes in thin layers, they are not taken into consideration for preventing the wave-guiding effect.

10 **SUMMARY OF THE INVENTION**

~~The invention is based on the object of overcoming the disadvantages of the prior art. In particular, the intention~~
One object of the present invention is to
15 provide an organic active OLED layer and a method for producing it in which particular precautions are taken to minimize the wave-guiding effect without the service life of the layer being severely shortened as a result of the use of organic materials or a high degree of wavelength selectivity of the materials.
20 ~~extraction occurring as a result of periodic structures.~~

~~This object is achieved by means of an OLED having the features of claim 1 and by means of a method having the~~
25 ~~features of claim 9. Advantageous developments of the OLED and of the method are specified in subclaims 2-8 and 10-16, respectively.~~

This and other objects are attained in accordance with one aspect of the present invention directed to an
30 organic light emitting diode (OLED) with at least one organic layer which has refractive index inhomogeneities, wherein the organic layer has at least one first partial region and at least one second partial region which comprise organic material and have
35 different refractive indices, and the partial regions form a layer with a composite-like structure.

Another aspect of the present invention is directed to a method for producing an organic light emitting diode (OLED) with at least one organic layer which has refractive index inhomogeneities wherein the material
5 of the organic layer is applied to a carrier in such a way that, during or after the coating step, at least one first partial region and at least one second partial region form in the layer. The partial regions have different refractive indices, and the partial
10 regions form a layer with composite-like structure.

According to an aspect of the invention, inhomogeneities are introduced in at least one organic layer of the OLED, said inhomogeneities themselves
15 having an organic composition. The organic inhomogeneities have a different refractive index than a layer matrix and consequently disturb the wave-guiding effects.

20 Figure 2A uses the line 4a provided with arrows to diagrammatically show the path of a photon which is emitted by an emitter 6 and extracted from an organic layer 1 when, after passing through partial regions 1B having a first refractive index within a layer matrix
25 material 1A having a second refractive index, different from the first, it impinges on a transparent adjacent layer 2, which is an ITO anode, for example, at a steeper angle than the critical angle of total reflection. Layer 5 is a cathode, for example.

30 For comparison, Figure 2B uses the line 4b provided with arrows to diagrammatically show the path of a photon which is generated in a homogeneous organic layer composed entirely of a single material. Between a
35 cathode 5 and a transparent anode 2, a photon remains trapped in the layer as a result of the wave-guiding effect if the angle of impingement does not exceed the

critical angle of total reflection. Layer modes are produced.

5 In order to produce an organic layer with partial regions with different refractive indices, use is made of a mixture of plastics, the properties of which permit utilization as an active OLED layer or else as a passive intermediate layer, or a mixture of its starting materials. Separation processes during or
10 after layer formation are utilized in a targeted manner in order to obtain a layer containing two or more phases in this case. In a particular preferred variant, the phases with a different refractive index comprise different plastics.

15 In order that the refractive index inhomogeneities contribute particularly effectively to the extraction of the photons from the transparent layer, it is advantageous that a composite-like structure forms as a
20 result of the separation in the layer. In this case, composite-like structure is understood as principally the structure of a particle-filled plastic, and also the structure of a composite with two three-dimensionally interpenetrating individual components,
25 as is also produced for example when an open-pored plastic body is filled with a second plastic.

In order to obtain a layer having at least two phases, at least two polymers are dissolved or dispersed, which
30 separate during the removal of the solvent or dispersant or already separate before the drying of the layer as a result of separation of the solvents or dispersants.

35 Instead of working with substances that have already been polymerized, it is conceivable to use monomeric or oligomeric starting materials of the polymers for coating, in which case the separation of the at least

two phases may occur before, during or after polymerization.

5 In another preferred variant, the layer is composed chemically of a single polymeric material which has, in island regions with respect to the matrix, differences in material properties such as crystallinity, degree of branching, degree of crosslinking, density and copolymerization, and thus differences in refractive
10 index.

Different charge carrier transport materials, emitter materials and arbitrary mixtures thereof may be used as plastics. It is also possible to use further,
15 electrically inactive plastics or their precursors.

By varying the production conditions, separation process and chemical reactions and thus the structure and the optical properties of the plastic layer can be
20 influenced in a targeted manner.

~~The invention is explained in more detail below using two exemplary embodiments in conjunction with figures 1 to 3.~~

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~~In the figures:~~ **BRIEF DESCRIPTION OF THE DRAWINGS**

Figure 1 shows a diagrammatic illustration of the waveguidance losses in the layers of a conventional organic light emitting diode,

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Figure 2A shows a diagrammatic illustration of the scattering of photons by refractive index inhomogeneities in an organic layer,

35 Figure 2B shows a diagrammatic illustration of the waveguidance in an organic layer without scattering centers,

Figure 3A shows a micrograph of an organic layer in accordance with a first exemplary embodiment of the invention,

5 Figure 3B shows a micrograph of an organic layer in accordance with a second exemplary embodiment of the invention.

10 **DETAILED DESCRIPTION OF THE DRAWINGS**

The explanations concerning Figures 1-2B are already given in the general part of the description.

Exemplary embodiment 1 (Figure 3A):

15 This involves the polymer materials poly(paraphenylene-vinylene) derivative (PPV) and poly(N-vinylcarbazole) (PVK), which were mixed and spun-on in solution as a layer on a substrate.

20 The mixture of the dissolved plastics comprised one third PPV and two thirds PVK. After the spinning-on, a separation of the two materials occurred, in the course of which large structures made of PVK formed with small satellites in a PPV matrix. This is shown in the
25 micrograph of the surface of the organic layer produced, as shown in Figure 3A. PVK regions with a broad size distribution curve are incorporated as scattering centers in a matrix made of PPV.

30 **Exemplary embodiment 2 (Figure 3B):**

As in exemplary embodiment 1, this involves PPV and PVK which were mixed and spun on in solution as a layer.

In this example, the mixture comprises 50% PPV and 50%
35 PVK. The micrograph of the organic layer produced in this case, illustrated in figure 3B, shows that spherical PVK regions are again incorporated as scattering centers in a PPV matrix, but in this case

with a very much narrower size distribution curve than in the first exemplary embodiment explained above.

5 In both the micrographs, the light areas are composed of PVK and the dark areas are composed of ~~PVV~~-PPV.

10 The exemplary embodiments verify how a different size and a different size distribution of the scattering centers can be obtained by varying the mixture ratios of the two plastics. This advantageously affords the possibility of setting the optical properties of the organic layer in a targeted manner.

15 It goes without saying that the present invention is not restricted to the polymer materials mentioned in the two exemplary embodiments, but rather can be applied to all materials suitable in terms of their electrical properties for organic LEDs.

20 Furthermore, with a single plastics material, for example, it is possible to obtain a similar effect to a separation if crystalline regions form within an amorphous matrix.

25 The scope of protection of the invention is not limited to the examples given hereinabove. The invention is embodied in each novel characteristic and each combination of characteristics, which includes every combination of features which are stated in the claims,
30 even if this combination of features is not explicitly stated in the claim.

~~Abstract~~ ABSTRACT OF THE DISCLOSURE

~~Organic light emitting diode (OLED) and method for producing it~~

An organic layer (1) is applied to a transparent carrier (2) in such a way that different partial regions with different refractive indices are formed in the layer. Owing to deflection at the phase boundaries within the layer, fewer photons remain trapped in the layer as a result of wave-guiding losses than in the case of homogeneous layers.

~~Figure-2A~~